

Probing a liquid to glass transition in equilibrium

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We use computer simulations to investigate the static properties of a simple glass-forming fluid in which the positions of a finite fraction of the particles has been frozen in. By probing the equilibrium distribution of the overlap between independent configurations of the liquid, we find strong evidence that this random pinning induces a glass transition. At low temperatures, our numerical findings are consistent with the existence of a random first order phase transition rounded by finite size effects.

Experiments allow to measure the increase of the viscosity of supercooled liquids approaching the glass transition over more than 15 orders of magnitude. Despite this large range, there is at present no consensus whether this phenomenon is controlled by an underlying phase transition, or whether relaxation times progressively increase down to zero temperature [1]. There is not even agreement on the microscopic mechanisms at work, despite the fact that glassy dynamics is commonly observed in a large variety of materials, from simple liquids, network forming liquids, to soft and biological matter [2]. Because supercooled liquids inevitably fall out of equilibrium at the experimental glass temperature T_g , amorphous solids form without encountering any singularity.

The existence of a thermodynamic transition at a finite temperature $T_K < T_g$ is highly debated [2]. Such a singularity was discussed long ago by Kauzmann [3] who pointed out that experimental data for the configurational entropy extrapolate to zero at T_K , the “Kauzmann temperature”. Physically, this would imply that for $T > T_K$ there exist exponentially many (in the number of particles) equilibrium states (neglecting vibrations), whereas below T_K this number becomes sub-exponential and the system forms an “ideal” glass. This view is supported by the analysis of the temperature dependence of the relaxation dynamics, which seems to be consistent with the existence of a temperature $T_0 > 0$ at which the relaxation time diverges [4]. This makes sense if thermodynamic and dynamical singularities occur at the same temperature, $T_0 \approx T_K$, but so far T_0 and T_K have been determined only by empirical fitting formula and uncontrolled extrapolations of the behavior of macroscopic observables. These procedures evidently leave much room for debates on the existence of such a transition [5, 6].

The existence of a true phase transition is appealing from a theoretical perspective, since a number of analytical results obtained within various approximate schemes or idealized limits suggest the existence of a “random first order transition” (RFOT) occurring in glass-forming materials [7, 8]. This transition results from the temperature evolution of a complex free energy landscape characterized by a large number of metastable states. It provides an exact realization of Kauzmann’s entropy crisis. However, the mean-field character of the RFOT, the technical difficulties posed by finite-dimensional fluctuations [9], and the indirect connections between theory

and experiments explain why RFOT is only one among several theoretical scenarios for the glass transition [2].

In the following we demonstrate that it is indeed possible to perform a detailed *equilibrium* study of a liquid-glass transition whose nature is equivalent to the one possibly occurring at T_K in bulk glass-formers [10, 11]. This allows us to probe numerically, for the first time, the nature of the phase transition and the microscopic properties of the glass phase at thermal equilibrium. In particular, we show that glass formation can be located without using uncontrolled extrapolations, and that equilibrium glasses can be produced.

We consider a 50:50 binary mixture of harmonic spheres [12, 13] of diameter ratio 1.4 at constant density $\rho = 0.675$, which we study using molecular dynamics (see SM for methodological details). This quasi-hard sphere system has an onset temperature around $T_{\text{on}} \approx 10$, and a mode-coupling temperature $T_{\text{mct}} \approx 5.2$ [13]. All numbers are expressed in appropriate reduced units (see SM). To efficiently sample the equilibrium thermodynamic properties of the system, we use replica exchange molecular dynamics [14]. We carefully ensure thermalization and equilibrium sampling by requiring that all particles move several times across the entire simulation box for each state point and that all replicas properly explore the configuration space.

Our central idea is to induce a glass transition by increasing the strength of a random pinning field in a thermalized dense liquid [10, 11]. We first equilibrate the liquid at a given temperature T , before freezing permanently the position of a certain set of particles. Among various other pinning geometries [11, 15, 16], here we randomly select a finite concentration of particles, c . Working at constant total number density ρ , we then study the equilibrium properties of the model in the (c, T) phase diagram. For each state point, we average the results over independent realizations of the random pinning field [15]. Intuitively, pinned particles constrain the available phase space of the remaining fluid particles, thus impacting their static and dynamic properties. Indeed, we find that the dynamics slows down dramatically with c at constant T , see Fig. SM1, in agreement with earlier studies [11, 17–19].

Freezing particles at the positions they occupy at equilibrium is a key ingredient to this study, since this does not perturb the interaction part of the Hamiltonian, but only constrains the available phase space. Indeed, any

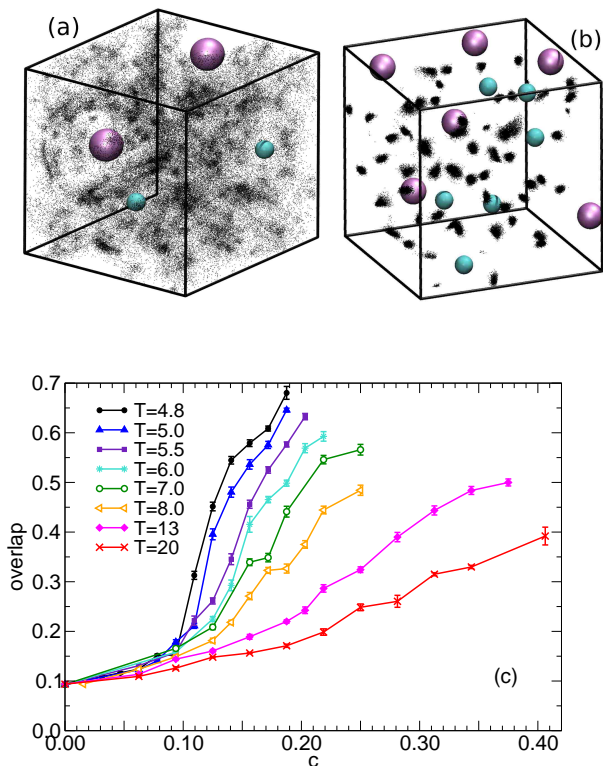


FIG. 1: (a) and (b): Large spheres represent pinned particles (rescaled in size by a factor 0.5), small dots are the superposition of the position of fluid particles obtained from a large number of independent equilibrium configurations at $T = 4.8$ for $c = 0.0625$ (a) and $c = 0.1875$ (b). The total number of particles is $N = 64$. Panel (c): Average overlap $\langle q \rangle$ as a function of concentration c of pinned particles for $N = 64$ and different temperatures.

ensemble averaged correlation function measured in the pinned liquid takes the same value as in the bulk system at $c = 0$ [15, 20]. Theoretical studies have recently revealed two additional consequences [10, 21]. First, increasing c affects the configurational space in the same way as decreasing temperature does for the bulk liquid [10]. In particular, the RFOT occurring at T_K^0 for $c = 0$ becomes a transition line $T_K(c) \geq T_K^0$ with equivalent properties. Second, the pinning field only induces a glass transition if temperature is low enough so that metastable states and glassy dynamics are well-developed at $c = 0$. The glass transition line $T_K(c)$ ends at a second order critical point [10], and no transition exists for temperatures above this point. Note that other types of pinning fields [22–24] produce qualitatively different results, because they perturb the Hamiltonian and possibly affect the nature of the glass transition [21].

In Fig. 1 we provide a qualitative illustration of our strategy. We show how an equilibrium glass state is obtained at constant temperature, $T = 4.8$, if the concentration c of pinned particles is increased. In these snapshots the large spheres are the pinned particles in

a typical realization of the disorder whereas the small black dots stem from the superposition of a large number of independent, equilibrium configurations visited by the fluid particles. For $c = 0$, the dots look like mist homogeneously filling the simulation box. For $c = 0.0625$, panel (a), the fluid particles are not much constrained and still have access to a large number of distinct configurations. Thus, the dots form fuzzy clouds. An increase to $c = 0.1875$, panel (b), condenses the dots into well-defined patches, which represent the highly constrained positions occupied by the free particles. This qualitative observation illustrates that increasing c leads to a *collective localization* of the fluid particles. We emphasize that during the simulations, all fluid particles diffuse and explore the entire simulation box. However, particles move in such a way that collective density fluctuations are frozen. Thus, the system is in a glass state characterized by a frozen amorphous density profile [2], but which can nevertheless be studied in equilibrium conditions because our replica exchange simulation algorithm permits single particle diffusion.

We now study in more detail how the system makes the transition between the fluid and glass states shown in Fig. 1. The images suggest that the number of available states, and thus the configurational entropy, considerably decreases with c . A quantitative determination of the configurational entropy is however difficult and has several shortcomings [2]. Therefore we use a microscopic order parameter to characterize the transition from fuzzy clouds to small patches seen in Fig. 1. An appropriate quantity is the overlap $q_{\alpha\beta}$ measuring the degree of similarity between two arbitrary configurations α and β , which has been used in spin glass models displaying an RFOT [7]. In practice, we discretize space into small cubic boxes of linear size 0.55, and define $n_i^{(\alpha)} = 1$ if box i in configuration α is occupied by a particle, and $n_i^{(\alpha)} = 0$ if not. Then,

$$q_{\alpha\beta} = \frac{1}{N_b} \sum_{i=1}^{N_b} n_i^{(\alpha)} n_i^{(\beta)}, \quad (1)$$

where the sum runs over the N_b boxes which do not contain pinned particles. By definition $q_{\alpha\alpha} = 1$, while a small overlap is obtained for independent configurations ($q_{\text{rand}} \simeq 0.11$ on average for an ensemble of independent configurations). In the remainder of the paper, we characterize the transition through a detailed analysis of the statistical properties of the overlap for a broad range of control parameters.

In Fig. 1c, we show the c -dependence of the average overlap, $\langle q \rangle = \langle q_{\alpha\beta} \rangle$, where the brackets stand for thermal and disorder averages. Above the onset temperature, $\langle q \rangle$ increases gradually with c . For $T \lesssim 8.0$, the growth remains modest at low c , but this initial regime is followed by a rapid increase in the range $\langle q \rangle \approx 0.25 - 0.4$. Finally, if T is decreased even further this rapid growth occurs for lower values of c and becomes sharper. At $T = 4.8$ it is sufficient to reach $c \approx 0.11$ to abruptly localize the fluid particles, $\langle q \rangle > 0.5$, while such a large

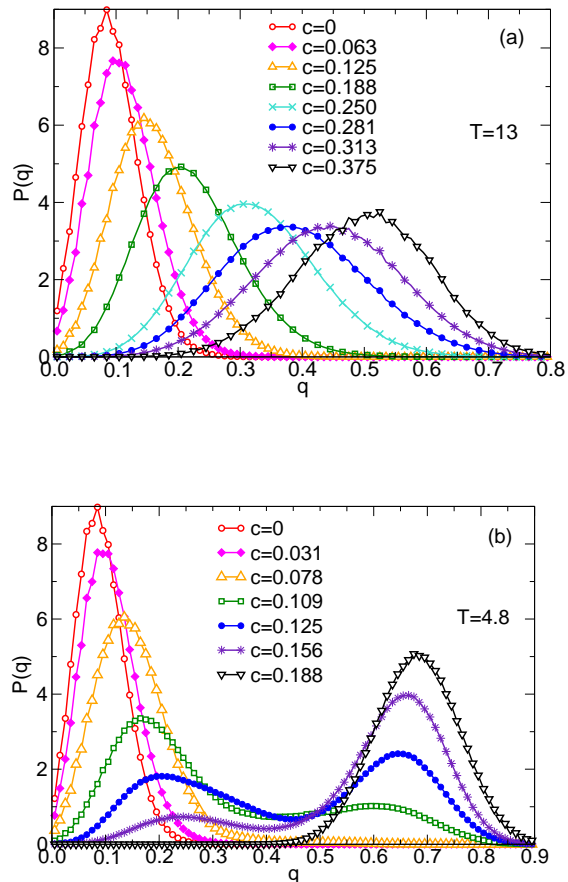


FIG. 2: Probability distribution of the overlap, Eq. (2), for different values of the concentration of pinned particles at high and low T (panels (a) and (b) respectively). Note the presence of a double peak structure at low temperatures.

overlap requires three times as many pinned particles, $c \approx 0.35$, near the onset temperature.

These observations show that at low temperatures the glass state is reached at a sharply defined c -value, whereas pinning acts quite smoothly at high T . The T -dependence of $\langle q \rangle$ is consistent with the emergence of a discontinuous jump at low temperatures, $T \lesssim 8.0$, but the relatively small system size shown in Fig. 1c, $N = 64$, will obviously smear out such a discontinuity. To overcome this difficulty we have determined the fluctuations of the overlap and in Fig. 2 we show the overlap distribution function,

$$P(q) = \langle \delta(q - q_{\alpha\beta}) \rangle. \quad (2)$$

At high temperatures, $T = 13$ in Fig. 2a, $P(q)$ evolves smoothly from distributions peaked at small q for small c to distributions peaked at large q values at large c . This is consistent with $P(q)$ becoming a delta-function for $N \rightarrow \infty$ for all c , and with the smooth increase of

$\langle q \rangle = \int_0^1 P(q)q dq$ at high T shown in Fig. 1c. A qualitatively different behavior is observed at low T , see Fig. 2b. A narrow peak is still present for small and large c , but $P(q)$ is *bimodal* for intermediate c . The presence of two peaks implies that it is equally probable that two independent thermalized configurations are either very similar or very different. The former situation is favored at larger c because too few distinct configurations exist, while the latter holds at small c when the pinning field is not strong enough to prevent the system from exploring a large configuration space. Bimodal distributions of the order parameter can be interpreted as the phase coexistence between the low- q liquid and the high- q glass phases, and the two-peak structure of $P(q)$ is suggestive of a first-order transition for the order parameter $\langle q \rangle$, rounded by finite size effects.

We emphasize that the above conclusions do not result from any kind of extrapolation, but follow directly from equilibrium measurements of a microscopic order parameter on *both* sides of the transition. To estimate more precisely the location of this putative transition, we have measured various moments of the distribution $P(q)$. From the second moment $\langle q^2 \rangle = \int_0^1 P(q)q^2 dq$, we define the static susceptibility,

$$\chi(c, T) = N(1 - c) [\langle q^2 \rangle - \langle q \rangle^2]. \quad (3)$$

The curves presented in Fig. 3a show that at high temperatures χ has a mild c -dependence, whereas it develops a well-defined peak for $T \leq 7.0$ whose height and location respectively increases and shifts to smaller c if T decreases. The observed growth of the peak $\chi(c, T)$ at low T is a direct evidence of increasing static correlations in the bulk system [11, 25]. The location of this peak allows to estimate the value of the critical concentration at the given temperature. As demonstrated in Fig. 3b, an even more accurate location is obtained by considering the skewness $\gamma(c, T)$ of the distribution $P(q)$,

$$\gamma = \frac{\langle (q - \langle q \rangle)^3 \rangle}{\langle (q - \langle q \rangle)^2 \rangle^{3/2}}, \quad (4)$$

since γ crosses zero when the distribution is symmetric, i.e. precisely at coexistence. From the location of the peak in $\chi(c, T)$ and the zero-crossing point of $\gamma(c, T)$, we can locate the fluid-glass coexistence line, as shown in Fig. 3c. These two quantitative estimates become reliable if $T \lesssim 8.0$, i.e. when bimodal distributions are present, and we see that they agree with each other within error bars (estimated using the jack-knife method). In the context of Fig. 3c, the existence of a Kauzmann temperature at $c = 0$ would rely on extrapolating our finite c transition points (determined without extrapolation) to the limit $c \rightarrow 0$.

Altogether, our results show that a glass transition is induced by random pinning, with nontrivial thermodynamic signatures occurring at a sharp value of the random pinning field that are consistent with the existence of an equilibrium random first-order transition [10]. However, since our results have been obtained for a rather

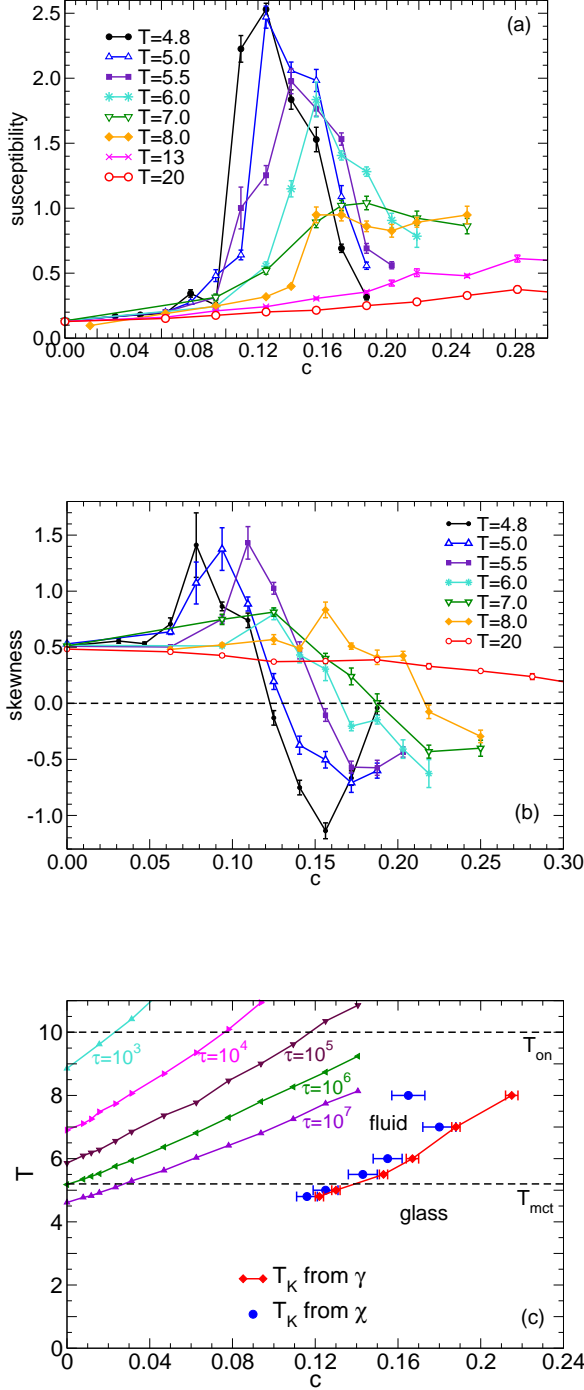


FIG. 3: (a) The static susceptibility χ , Eq. (3), as a function of concentration c for different temperatures. (b) c -dependence of the skewness γ for different T . (c) The equilibrium phase diagram determined from the position of the peak in the susceptibility χ and from the root of the skewness γ showing the location of the fluid and glass phases. Also included are curves of constant relaxation time as determined from the self-intermediate scattering function.

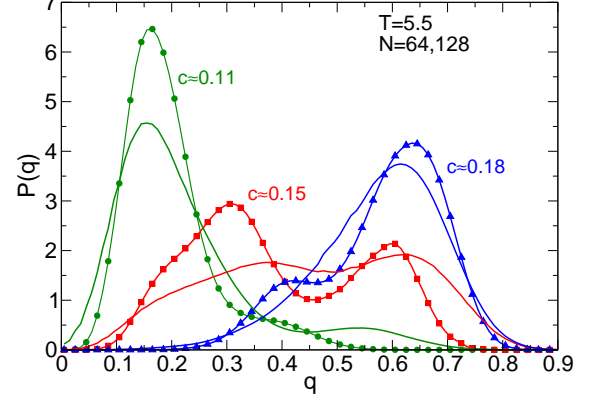


FIG. 4: System size dependence of the overlap distribution function for $T = 5.5$ and three values of c below (green), near (red), and above (blue) the transition for $N = 64$ (lines) and $N = 128$ (lines with symbols). The peak susceptibility increases from $\chi \approx 1.8$ for $N = 64$ to $\chi \approx 2.7$ for $N = 128$ (data not shown).

small system size, $N = 64$, they do not establish its existence in the thermodynamic limit. Unfortunately, the parallel tempering algorithm we use becomes less efficient when N is increased. While we cannot access very low temperatures for larger N , we can, however, investigate the N -dependence in a limited range. Our results for larger systems confirm that no sharp transition exists for $T \geq 8.0$, while we obtained clear signs of enhanced bimodality and static susceptibility when $T < 6.0$, as shown in Fig. 4 for $T = 5.5$. The small dip in $P(q)$ for $N = 64$ and intermediate c values becomes more pronounced for $N = 128$, while the peaks at low and large overlaps become sharper. Such system-size dependence is again typical of first-order transitions. A more extensive finite-size scaling analysis would be useful, but it is at present beyond the computational means.

In contrast to conventional studies of the glass transition, our approach allows us to study both liquid and glass phases at thermal equilibrium, it does not require uncontrolled extrapolations, and we characterize the glass formation by means of a microscopic order parameter. Our work thus demonstrates the feasibility of systematic equilibrium studies of the nature of the liquid-to-glass transition. Our approach is very general and can easily be applied to any type of glass-former. Also, by using optical tweezers it will be possible to use this method to study colloidal glasses and thus to produce experimentally what is often considered as “impossible-to-reach ideal glass states” [26]. Therefore our work opens the door for a new generation of direct, systematic studies of the nature of the glass state and for the production of novel amorphous materials.

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Supplementary Material

I. MODEL AND DETAILS OF THE SIMULATIONS

The model we have been using is a 50:50 binary mixture of N harmonic spheres [1]. All particles have the same mass m and particles i and j separated by a distance r_{ij} have an interaction given by

$$V(r_{ij}) = \frac{\epsilon}{2}(1 - (r_{ij}/\sigma_{ij})^2) \quad . \quad (5)$$

Here $\sigma_{11} = 1.0$, $\sigma_{12} = 1.2$, and $\sigma_{22} = 1.4$. We will use σ_{11} as the unit of distance, $\sqrt{m\sigma_{11}/\epsilon}$ as the unit of time and $10^{-4}\epsilon$ as unit of energy, setting the Boltzmann constant $k_B = 1.0$. The density we have been using is 0.6749715.

Before we discuss the details of the procedure how we have pinned the particles, we recall that pinning particles in a liquid configuration that has been equilibrated at a temperature T_{pin} does not perturb the equilibrium properties of the remaining liquid at the temperature $T_{\text{liq}} = T_{\text{pin}}$ if one averages over a sufficiently large number of realisations of pinned configurations [2–4]. Therefore it is not necessary to re-equilibrate the pinned system after the pinning. If instead T_{pin} is different from the simulation temperature T_{liq} of the liquid the resulting Kauzmann line $T_K(c, T_{\text{liq}})$ will depend on T_{pin} as well [5].

To prepare the system at different concentrations c of pinned particles we have proceeded as follows. First we have used a standard molecular dynamics simulations to equilibrate a liquid (in the bulk) at the target temperature T_{liq} . For this we have integrated the equations of motion using the velocity form of the Verlet algorithm with a time step of 0.02. Let us call one of the so obtained bulk configurations C_{bulk} . Subsequently we have to determine which ones of the particles have to be pinned. Although choosing these pinned particles randomly from the C_{bulk} configuration is certainly a possibility, such a choice can lead to systems in which many pinned particles are clustered or that have relatively large regions in which there are no pinned particles. These fluctuations make that the transition at the Kauzmann point becomes smeared out at intermediate temperatures and hence makes it difficult to locate this transition accurately. Therefore we have chosen a scheme that makes that the typical distance between pinned particles, and therefore the size of

the remaining liquid regions, is relatively uniform. In practice we have generated an equilibrated liquid configuration of $c \cdot N$ particles, at density $\rho = 0.672$, using a molecular dynamics simulation with periodic boundary conditions. Let us call a typical configuration of this simulation C_{pin} . Since this is a liquid like configuration, all particles have a similar number of nearest neighbors and also a well defined nearest neighbor distance from each other. Subsequently we have scaled this configuration such that the box size becomes the same as the one of configuration C_{bulk} . Then we have searched for each particles of the C_{pin} configuration the closest particle in the C_{bulk} configuration. These closest particles have then been pinned permanently. In short this procedure allows to select from the C_{bulk} configuration $c \cdot N$ particles that can be pinned permanently and that have a relatively uniform distance from each other.

Typically we have averaged our results over around 5 independent realisations of the pinning field if c was small and over up to 20 realisations when c was large, i.e. around the Kauzmann point. The total amount of CPU time used for obtaining the presented results is about 900,000 hours.

II. RELAXATION TIMES

In order to determine the relaxation time of the pinned system as a function of temperature and concentration c we have used standard molecular dynamics simulations with a system of $N = 1000$ particles (step size 0.05). From these runs we have determined the self intermediate scattering function $F_s(q, t)$, where q is the wave-vector. Using for q the value of 5.5, which corresponds to the position of the maximum in the static structure factor, we have defined the relaxation time τ via $F_s(q, \tau) = e^{-1}$. In Fig. 5 we show the temperature dependence of $\tau(T, c)$ in an Arrhenius plot. From this graph one recognizes that for $c = 0$, i.e. the bulk system, the data curves slightly upwards, i.e. the system is a somewhat fragile glass-former, as discussed in Ref. [1]. An increase of c has two effects: 1) The dynamics is slowing down very strongly (by several orders of magnitude at intermediate and low T) and this slowing down becomes more pronounced if T is decreased. 2) At the highest c considered the relaxation dynamics becomes Arrhenius-like, i.e. the data falls on a straight line. This observation is in qualitative agreement with the theoretical prediction by RFOT [10] which shows that this aspect of the theory seems to be compatible with the simulation data. (We mention that a similar decrease of fragility with increasing c has been observed in Ref. [7].)

Since Fig. 5 demonstrates that the relaxation time is a strong function of c , one can expect that the relevant temperatures of the glass-former, such as the onset temperature T_0 , the mode-coupling temperature T_c , and the Kauzmann temperature T_K will also depend on c and will increase if c is increased. As a consequence it should

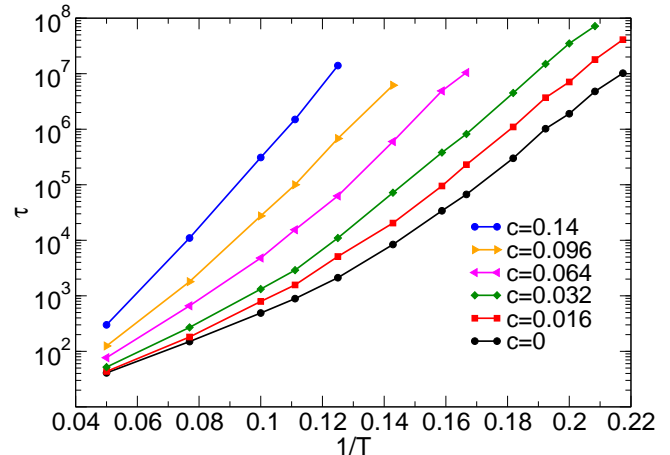


FIG. 5: Arrhenius plot of the relaxation time for different value of the concentration c of pinned particles.

become possible to cross the line $T_K(c)$ by doing equilibrium simulations at low T by increasing c [6, 8] and in the main text we show that this crossing, and hence the investigation of the occurring transition, can indeed be done in equilibrium.

Since the standard molecular dynamics slows down quickly upon approaching the $T_K(c)$ line, we have used the parallel tempering algorithm [9, 10] using 24 replicas. The smallest difference in temperature between two neighboring replicas was 0.3, which has allowed for a good overlap of the two potential energy distributions. Attempts to switch two neighboring replicas have been made every 50,000 time steps. We have checked that each parallel tempering run has indeed reached equilibrium by following any given replica and making sure that all temperatures have been sampled sufficiently. A typical path of a replica in temperature space is shown in Fig. 6 for our lowest studied temperature.

The parallel tempering algorithm does indeed allow to equilibrate the system down to $T = 4.8$ even if the concentration c of pinned particles is large. This can be recognized from Fig. 7 where we show the mean squared displacement (MSD) of the particles (distinguishing small and large particles) as a function of time. To calculate the quantity we have followed a given replica in temperature space and considered only time intervals at which this replica was at the target temperature T_{liq} . The figure shows that at sufficiently long times the MSD becomes very large, thus indicating that the particles do indeed move through the box also at high values of c , even if their *relative* arrangement does not change much, i.e. one is in the glass state. Or stated otherwise: Each particle explores all the small patches shown in Fig. 1a of the main text.

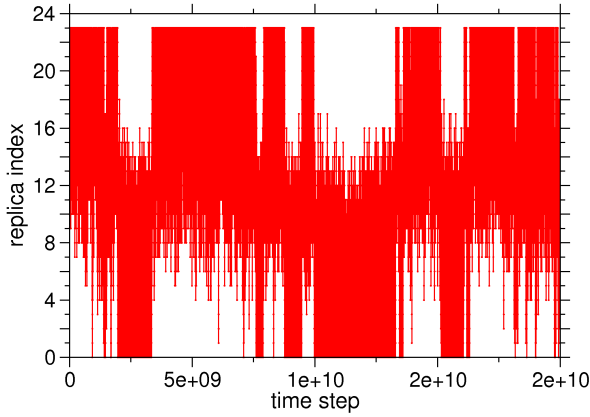


FIG. 6: Time dependence of a typical trajectory of a replica in temperature space. The temperatures are labeled 0-23, with 0 being the target temperature T_{liq} . The example shown is for $T_{liq} = 4.8$ and the concentration of pinned particles is $c = 0.14$.

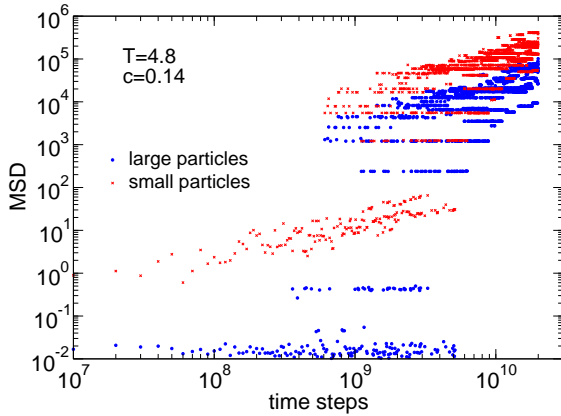


FIG. 7: Mean squared displacement of the particles as a function of time using the parallel tempering dynamics. The circles and crosses correspond to the large and small particles, respectively. $T = 4.8$ and $c = 0.14$.

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